Modeling to Support Fuel Development for an Advanced Fuel Cycle

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December 15, 2005

Advanced Simulations: A Critical Tool for Future Nuclear Fuel Cycles
Session 2F: Fuel Development

Lawrence Livermore National Laboratory
B543, R2074

OAK RIDGE NATIONAL LABORATORY
U. S. DEPARTMENT OF ENERGY

Modeling and Simulation will Benefit all Aspects of Fuel Development

Key Attributes for an Advanced Fuel Cycle:

Cm-242

- **Ability to Transmute TRU (Np-Pu-Am-Cm) will require:**
  - Remote fuel fabrication process
  - Material form to burn Am/Cm effectively
  - Separations process for An/Ln split
- **Recycle Plant Integration for Economy - to include:**
  - Co-location & integration of facilities (storage-separation-refabrication-waste)
  - Continuous, close-coupled, super-reliable, scalable processes
  - Optimum timing and staging of operations and deployment
  - Key Proliferation Resistance Features
  - VERY low losses, releases, waste, and interim storage
- **High Performance Fuel to improve overall Fuel Cycle metrics:**
  - High burnup
  - High fuel power-density and power-cycle temperatures
  - Large safety margins

High-Fidelity Simulation of Fuel Performance is the *Grand Challenge*

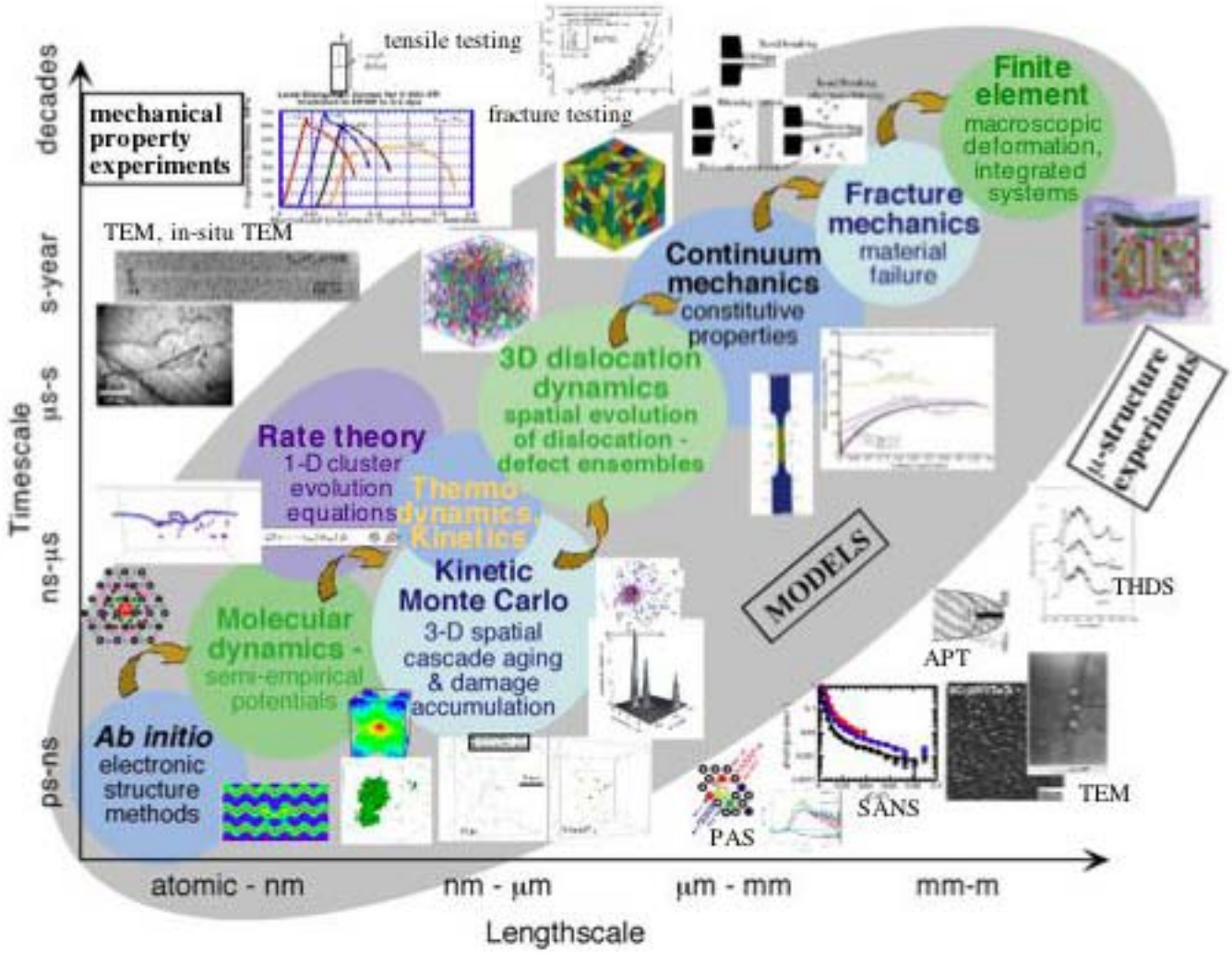
This Challenge requires an extended commitment from a multi-disciplinary team:

- It can build upon simulation for high-temperature materials and radiation-effects physics.
- It requires prediction of an evolving fuel chemistry and microstructure in a dynamic nuclear environment.

This is a worthy challenge, but no one can predict when the goal will be reached.

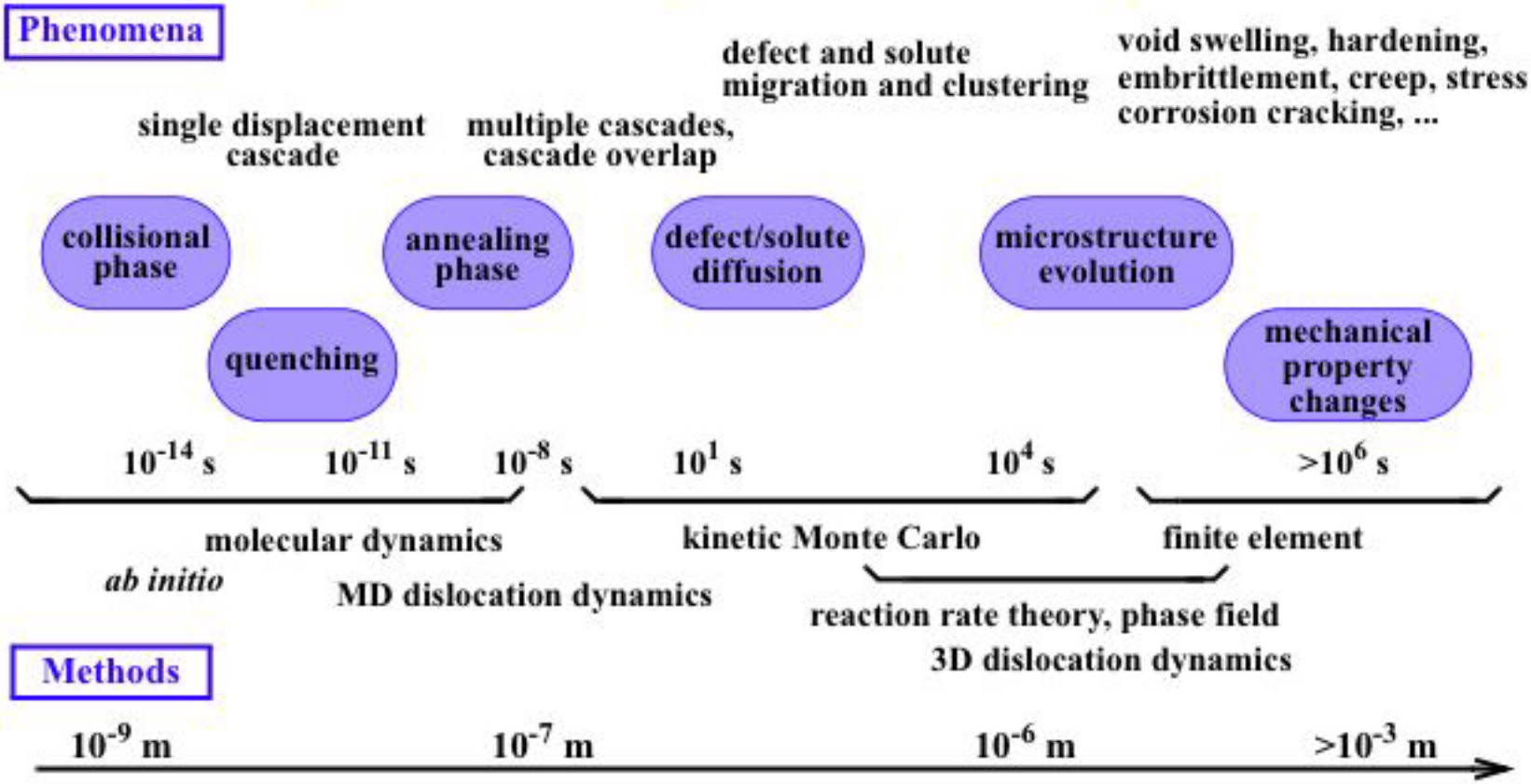
Can a near-term goal be accomplished along the way ?

Radiation damage is inherently multiscale with interacting phenomena ranging from ps to decades and nm to m



B.D. Wirth, UC-Berkeley

Schematic diagram: relevant phenomena and computational methods



Goal of multiscale modeling is fully predictive capability, but: "Prediction is very difficult, especially if it's about the future." ... Niels Bohr

There are Modeling and Simulation Benefits to be Gained in the Near-Term

The following areas need urgent attention:

Screening of candidate fuel materials:

primary thermodynamic evaluation
beginning-of-life fuel properties
evaluation for harmony within the fuel cycle
fabricability, recyclability

Evaluating the processes to make fuel

evaluating near-term viability, finding improvements

These areas are steps on the way to Fuel Performance Simulation, or are synergistic/precursor activities.

New Fuel Forms must be Evaluated against Constraints

Many researchers have been in an exploratory phase – looking for high-performance forms for burning TRU.

For near-term applications, focus on a few options in greater depth:

Choices will be made, and they should be informed by the information we can develop:

- some information can be drawn from experiments
- for Np, Am, and Cm the experimental evidence is scarce

We should explore the ability of modeling and simulation to help inform choices for near-term development.

Thermodynamic Analysis is the First Step

ORNL conducted a preliminary screening assessment of fuel matrices for coated-particle fuel containing TRU:

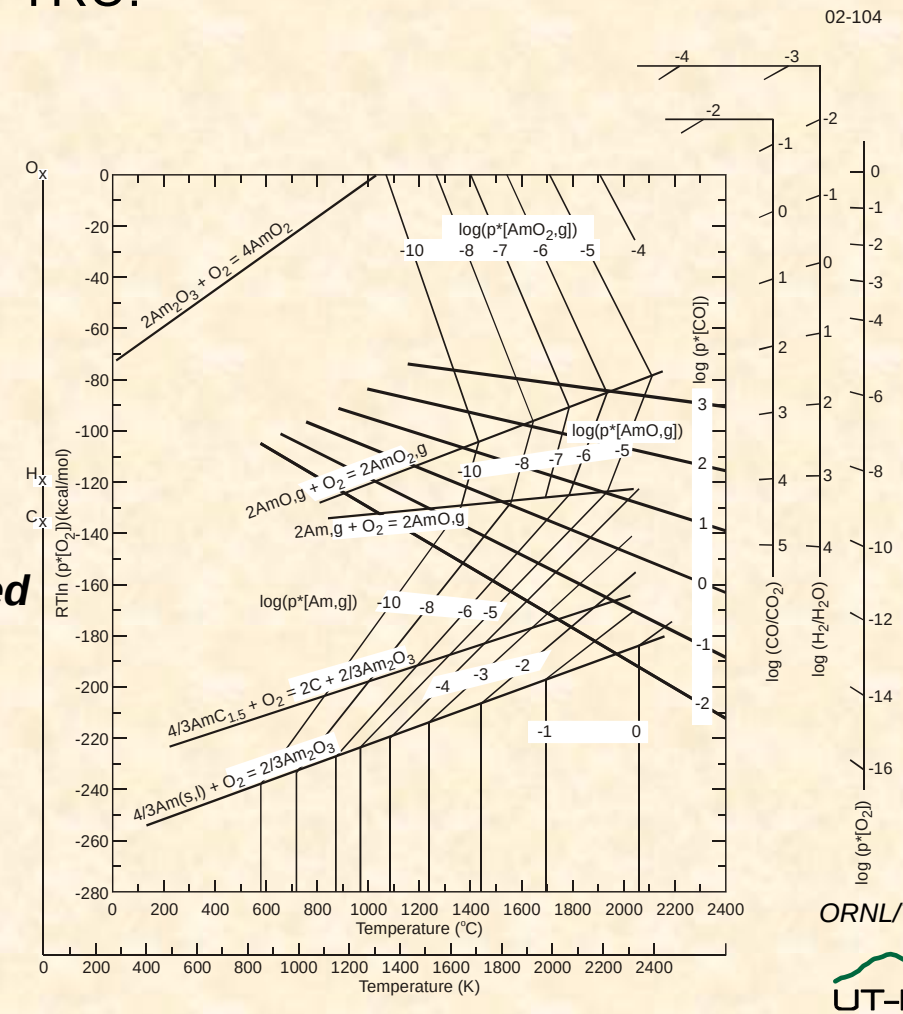
Am volatility was found to be:

low for oxides,

high for nitrides, carbides, and oxy-carbides,

and extremely high for metals.

Only oxides were suitable for coated particle fuels containing Am.



ORNL/TM-2002-133

The Production Process for a Number of Advanced Fuels Exhibit Key Uncertainties

Nitrides - Am volatility, material reproducibility and density/strength
Oxy-carbides ("UCO" for HTR): too variable in properties for an industrial process
Metallic fuels: Am-volatility must be addressed for plant-scale process
MgO-matrix - unexpected low-melting phase during reductive synthesis, uncertain retention of conductivity after irradiation. (GLOBAL 2005 #258, 448)

New Dispersion Fuels (CERMET, CERCER's): a thermodynamic analysis of new materials is needed for potential reactor performance and fuel cycle compatibility. It is likely that composite materials will find an application in fuels, as they have in materials.

Thermodynamic and Kinetic tools exist to help model the evolution of chemistry during the production process:

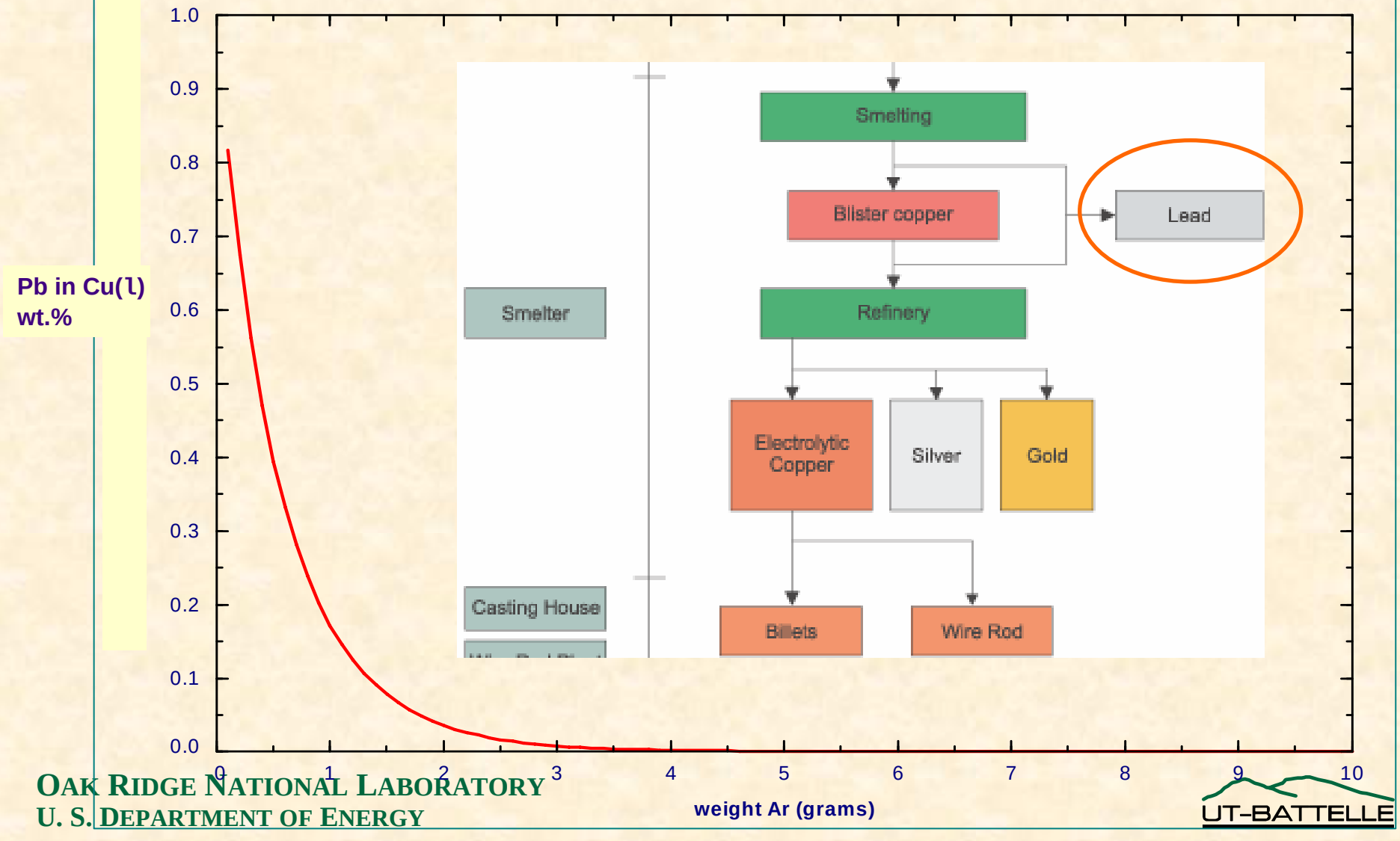
Thermodynamic *stream-calculations* in FACTSAGE, linked to kinetics in CHEMKIN, and transport in DICTRA.

Examples of Useful Stream-Calculations for Analysis of Materials Chemistry

Thermochemical Stream Calculation Provides Measure of Pb left in Cu as a Function of Total Inert Gas (Ar) Flowing Through Process

Deleading of Cu-1wt.%Pb by Argon Vacuum Refining

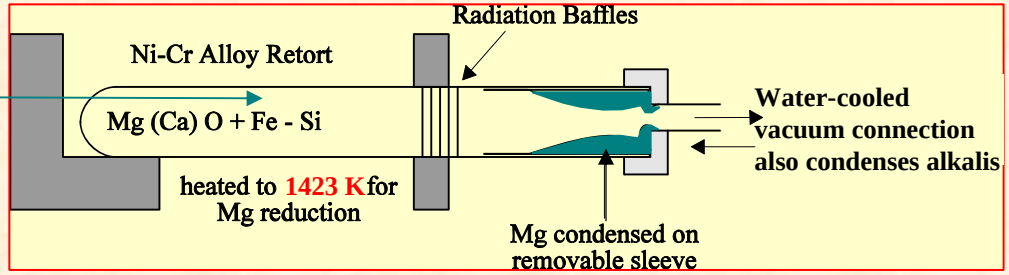
Example of «Open» Equilibrium Calculation: T=1200°C and P=0.001 atm.



Thermochemical Model of the Pidgeon Process for the Production of Magnesium

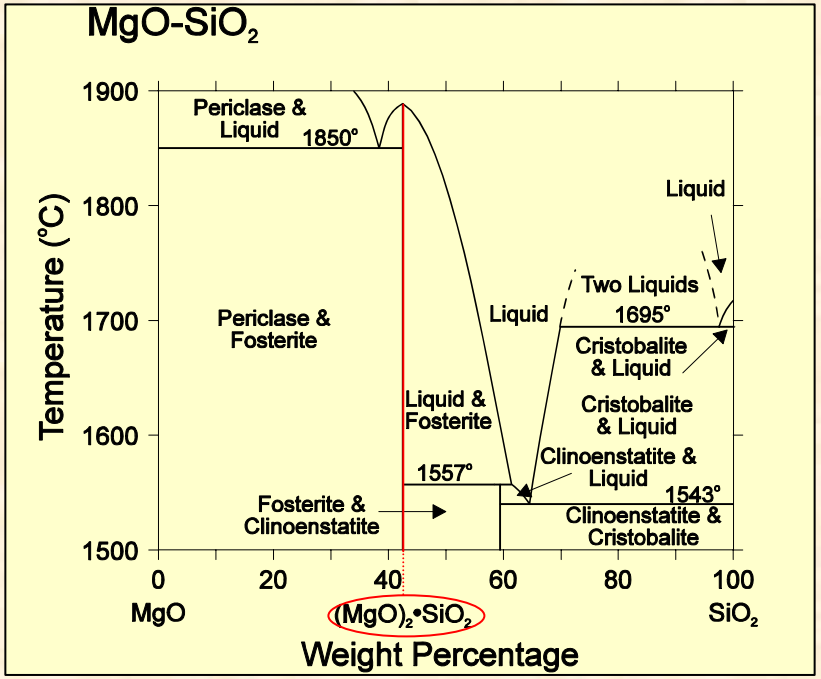
Apparatus Schema:

Equilibrium **Mg partial pressure** developed at the hot end of the retort



Calculations show magnesium production is enhanced by:

- reducing the total pressure (**<0.0010876 atm**);
- reducing a_{SiO_2} – this is done automatically due to $(MgO)_2 SiO_2$ formation, but the addition of say CaO (slag formation) reduces a_{SiO_2} further.



Note: MgO(s) and SiO_2(s) **cannot** coexist.

Although Oxide Fuels are Familiar, Science can make contributions to decisions regarding this proven system

Fuels for all types of reactors will likely be driven to higher burnup:

- The composition of phases in oxide fuel at these higher burnup levels is not known.

- More TRU elements will be produced at high burnup - and recycled.

- Detailed knowledge of oxygen transport in these fuels will be needed to adequately simulate fuel behavior. This same type of basic knowledge is needed for developing solid-oxide fuel cells.

- MgO matrix material needs further evaluation

In some instances a detailed model of a complex process is key to the success of a recycle plant

Key examples:

Fluidized-Bed Chemical Vapor Deposition (TRISO fuel)

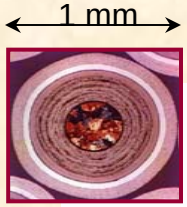
Casting for metal-fuels containing Am

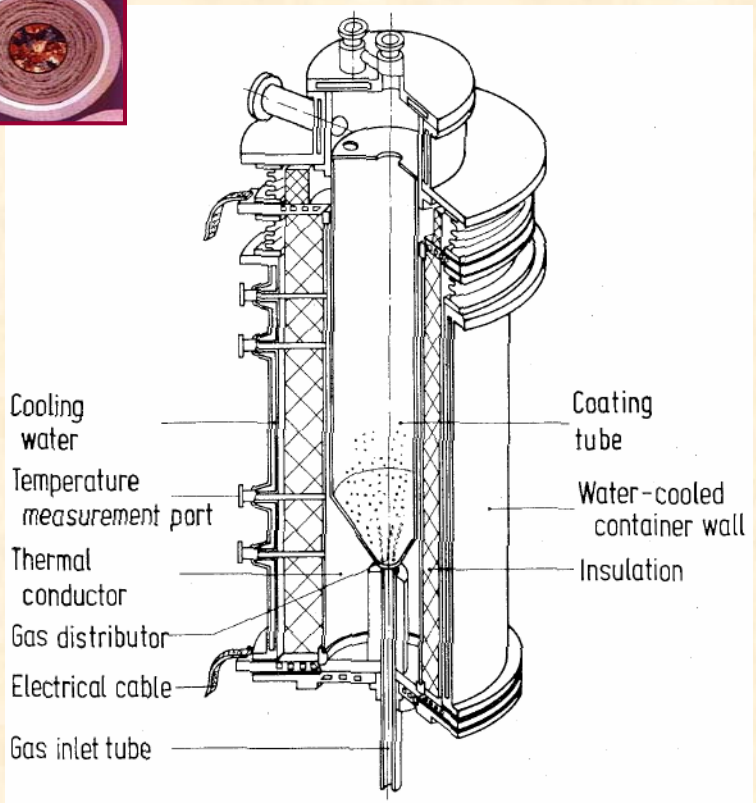
Co-Conversion linking Separations and Fuel Fabrication

Head-end process preparing spent-fuel for Separations

Voloxidation or other advanced head-end treatment

Fluid-Bed Processes will be necessary for an Advanced Fuel Cycle - Coating is a good Example





Simulation of solids velocity using MFIX code

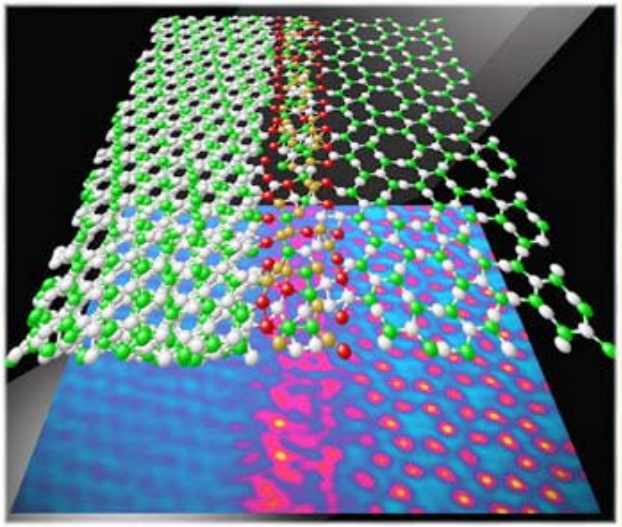
QuickTime™ and a
Microsoft Video 1 decompressor
are needed to see this picture.

2005 AIChE Meeting #301a

Coupled: Hydrodynamics, Reaction Kinetics, Heat and Mass Transfer, Particle Dynamics
All effect coating microstructure !

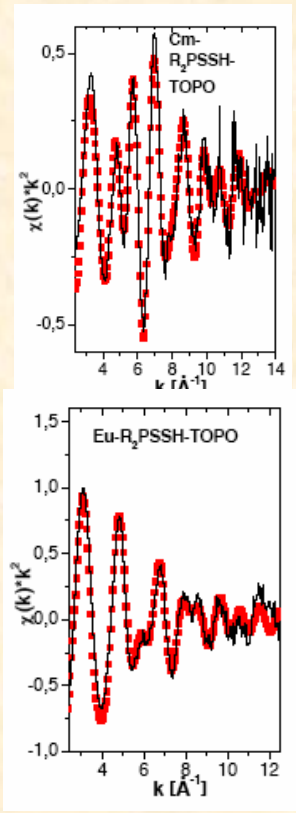
Nano-structural characterization of reference materials is necessary to validate the results of complex simulations

Fuel
materials



0.6 angstrom resolution of Silicon Nitride doped with La_2O_3

http://www.ornl.gov/info/ornlreview/v37_3_04/article15.shtml



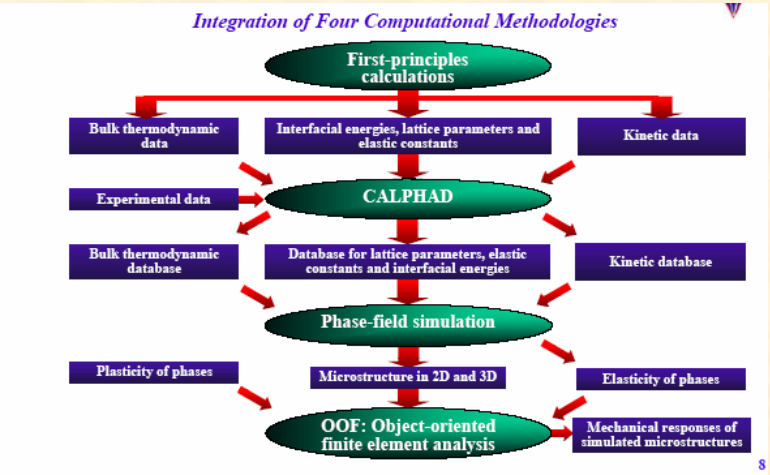
Separations
chemistry

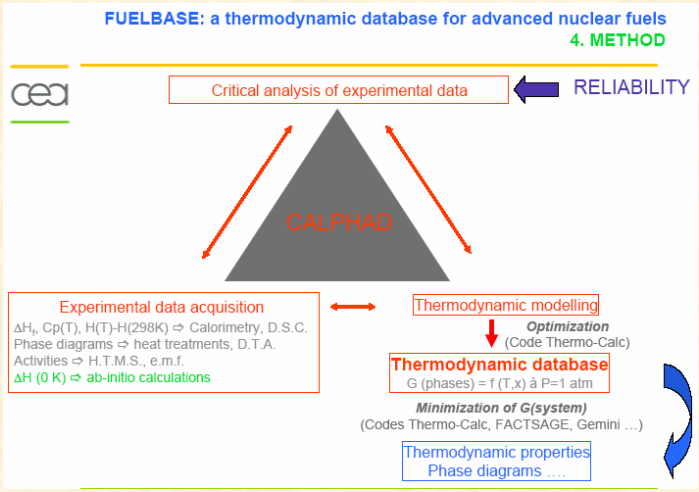
German comparison of EXAFS and TRLFS with quantum theory for candidate An/Ln extractants *GLOBAL 2005*

- We now have extraordinary tools for nano-characterization
- We also have archived reference materials that should be characterized to provide key validation of simulations.

Conceptual Models are needed to begin simulation of Fuels (as compared with ab initio simulation)

• Today, advanced materials are developed by a combined approach that draws the key information from various approaches (from MMSNF-4):





• Development of conceptual models allows more rapid progress and is a strategic investment

• We can also reap the benefit of heavy-element science performed over the past 40 years.

Modeling and Simulation can make near-term contributions to on the way to the accomplishing *the Grand Challenge*

- Candidate materials systems are screened based upon combined information available from experiments and modeling/simulation.

- Complex processes are analyzed and problems associated with bottlenecks or viability are quantified to support decisions.

- An optimized experimental program is designed to resolve key uncertainties and reduce development risk.

BACKUP

Overlap with Materials Simulation